



Research article

Field versus laboratory experiments to evaluate the fate of azoxystrobin in an amended vineyard soil

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ABSTRACT

This study reports the effect that adding spent mushroom substrate (SMS) to a representative vineyard soil from La Rioja region (Spain) has on the behaviour of azoxystrobin in two different environmental scenarios. Field dissipation experiments were conducted on experimental plots amended at rates of 50 and 150 t ha⁻¹, and similar dissipation experiments were simultaneously conducted in the laboratory to identify differences under controlled conditions. Azoxystrobin dissipation followed biphasic kinetics in both scenarios, although the initial dissipation phase was much faster in the field than in the laboratory experiments, and the half-life (DT₅₀) values obtained in the two experiments were 0.34–46.3 days and 89.2–148 days, respectively. Fungicide residues in the soil profile increased in the SMS amended soil and they were much higher in the top two layers (0–20 cm) than in deeper layers. The persistence of fungicide in the soil profile is consistent with changes in azoxystrobin adsorption by unamended and amended soils over time. Changes in the dehydrogenase activity (DHA) of soils under different treatments assayed in the field and in the laboratory indicated that SMS and the fungicide had a stimulatory effect on soil DHA. The results reveal that the laboratory studies usually reported in the literature to explain the fate of pesticides in amended soils are insufficient to explain azoxystrobin behaviour under real conditions. Field studies are necessary to set up efficient applications of SMS and fungicide, with a view to preventing the possible risk of water contamination.

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1. Introduction

Crop protection is an integral part of modern agriculture, with pesticide application being a major component. However, the presence of pesticides in soils and waters has increased in recent years (Herrero-Hernández et al., 2013; Köck-Schulmeyer et al., 2014; Masiá et al., 2015), and this practice has become extensively and hotly debated. Pesticide residues in the soil may be taken up by plants, degraded into other chemical forms, or washed into surface and ground waters. The leaching of pesticides into groundwater is a cause of concern, as this is one of the major sources of drinking water in many locations.

A positive step toward reducing pesticide leaching, and hence the risk of groundwater contamination by pesticide residues, involves enhancing the retention and degradation of pesticides in

soils and this can be achieved by increasing soil organic matter (OM) content. As it is known these processes depend on the soil OM content for the non-ionic hydrophobic pesticides (Marín-Benito et al., 2012a,b).

Nowadays, a common way of increasing the OM content involves soil amendment with organic residues (Moreno-Casco and Moral-Herrero, 2008). This soil amendment is used to increase OM content in Mediterranean agricultural soils, and there has recently been an increase in interest in assessing the application of these organic residues as a strategy for enhancing the retention and degradation of pesticides in soils in order to avoid their leaching into groundwater.

There are numerous studies researching the effect of organic amendments on the fate of pesticides in soils conducted under laboratory conditions (Fenoll et al., 2011; Martín et al., 2012; Rodríguez-Cruz et al., 2012a,b; López-Piñero et al., 2013), and although these studies provide valuable information on the effect organic amendments have on the behaviour of pesticides in the soil,

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they do not always reflect what actually occurs in the field. Consequently, both field studies and laboratory studies are needed to provide complete information on how pesticides dissipate under natural conditions. In general, there are few studies on dissipation in the field (Papiernik et al., 2007; Chai et al., 2009), with only a handful examining the influence of different organic amendments on pesticide dissipation (Dolaptsoglou et al., 2009; Herrero-Hernández et al., 2011a). Very few studies have evaluated the dissipation of pesticides under field and laboratory conditions simultaneously (Ahmad et al., 2003; Potter et al., 2005; Chai et al., 2013).

Azoxystrobin is a fungicide with a broad spectrum of systemic activity for the control of fungal crop pathogens (Bartlett et al., 2002). It belongs to the strobilurins group, and it is a chemical that has been approved for use on more than 80 different crops in 72 countries. Azoxystrobin is commonly used as a foliar fungicide, and a high proportion of this fungicide may be deposited in the soil when applied in spray form (Adetutu et al., 2008). Some reports on its persistence and mobility have indicated that it may remain in soils for several months (Bending et al., 2007). However, the presence of azoxystrobin detected in the ground and surface waters in different vine growing areas in Spain (Herrero-Hernández et al., 2013), Brazil (Menezes Filho et al., 2010), France (Rabiet et al., 2010), and Germany (Neumann et al., 2003) reveals that certain uncontrolled factors affect its behaviour in soils.

There are studies on the degradation of this fungicide in some soils amended with organic residues under laboratory conditions (Ghosh and Singh, 2009a; Sopeña and Bending, 2013), but the dissipation of azoxystrobin under field conditions has scarcely been studied (Gajbhiye et al., 2011), and to our knowledge there are no studies on the dissipation of azoxystrobin in soil amended with spent mushroom substrate (SMS). SMS is the composted organic material remaining after a mushroom crop has been harvested, and it is being generated on farms in increasing quantities (Martín et al., 2009). SMS is used as a soil fertilizer and amendment to increase the OM content of vineyard soils in La Rioja region (Spain).

The objective of this research was to study the effect that SMS soil amendment has on the fate of azoxystrobin under two environmental scenarios. Accordingly, field versus laboratory experiments were conducted for comparative purposes on a vineyard soil from La Rioja region, both unamended and amended with SMS at two rates, with the aim being to assess the dissipation, persistence, and mobility of azoxystrobin applied at two doses. In support to explain the fate of the fungicide, changes in adsorption of azoxystrobin by unamended and amended soil from field experimental plots and changes in the dehydrogenase activity of unamended and amended soil, untreated and treated with azoxystrobin in field and in laboratory were evaluated over the time.

2. Materials and methods

2.1. Chemicals and organic amendment

An analytical standard of azoxystrobin (Methyl (E)-2-[2-[6-(2-cyanophenoxy) pyrimidin-4-yloxy]phenyl]-3-methoxyacrylate) from Dr. Ehrenstorfer, Germany (99.0% purity) and the commercial formulation of azoxystrobin from Ortiva (Syngenta, Switzerland) were used in the laboratory and field experiments, respectively. Azoxystrobin is a fungicide with a water solubility of 6 mg L⁻¹ and a log Kow of 2.5 (Tomlin, 2000). All other chemicals used were supplied by Sigma–Aldrich Quimica SA (Spain).

Spent mushroom substrate was supplied by INTRAVAL Environmental Group TRADEBE S.L. (Spain). Its composition was described by Herrero-Hernández et al. (2011a) and their physico-chemical characteristics determined as described in this previous work are: pH 7.5, organic carbon (OC) content 27.1% and dissolved

organic carbon (DOC) content 1.22%.

2.2. Field experimental design and soil sampling

The field experiment was conducted in a vineyard in Sajazarra, La Rioja, Spain (42°35'0"N latitude and 2°57'0"W longitude). The soil used was a sandy clay loam soil classified as Typic Calcixerept. Their characteristics were determined by usual analytical methods (MAPA, 1986) (Table 1). Rainfall and temperature were recorded over the 378 days of experimentation at a weather station close to the study site (4 km east) (Fig. S1 in Supplementary material).

An experimental layout of randomized complete blocks (18 plots of 1.50 m × 3.90 m) was set up with six treatments (unamended (S) and amended soils (Soil + SMS) at two rates of SMS treated with two doses of azoxystrobin) and three replicates per treatment as indicated in Herrero-Hernández et al. (2011a,b). Unamended and amended soils (0–10 cm) at the rates of 50 or 150 t ha⁻¹ (Soil + SMS50 and Soil + SMS150) on dry weight basis were prepared on November 2010. Azoxystrobin solutions at two doses (0.25 and 1.25 kg ha⁻¹) were applied to the plots from the commercial formulation Ortiva-Syngenta (25% w/v of a.i.). Three more plots one unamended and two amended with 50 or 150 t ha⁻¹ of SMS, respectively, did not receive fungicide application (control plots). Soil samples from these untreated plots were collected for assays in the laboratory as indicated below.

Five topsoil samples were collected from 0 to 10 cm at 0, 2, 7, 14, 21, 28, 35, 84, 115, 150, 181, 235, 300 and 378 days after treatment to determine fungicide dissipation and five soil cores were collected to a depth of 50 cm after 84, 181 and 378 days of azoxystrobin application to determine mobility of fungicide. Soil samples were managed as indicated in Herrero-Hernández et al. (2011a).

Topsoil OM content was determined two days after SMS application and OM content at different depths of soil cores taken at 84, 181 and 378 days was also determined by triplicate. Results are included in Table 1 and Fig. 2.

2.3. Laboratory experimental design and soil sampling

Soil samples for laboratory experiments were taken from 0 to 10 cm of experimental plots prepared for fungicide dissipation study in field on November 2010. Soils collected from control plots, unamended and SMS-amended at dose of 150 t ha⁻¹, and without fungicide applied, were transferred to polypropylene containers. They were transported to the laboratory and treated as previously indicated for field soil samples.

Solutions of fungicide were prepared in sterile UHQ water and volumes of 10 mL of suitable concentrations to give similar concentrations to those applied in the field experiment (0.25 and 1.25 kg ha⁻¹) were added to 500 g fresh weight of soils to determine fungicide dissipation. Unamended and amended soils were then incubated in containers at 20 ± 2 °C in the dark during the experiment. The initial moisture content of the soils was adjusted to 40% of their maximum water holding capacity, and it was kept constant during the entire period of the experiment by adding sterile UHQ water when necessary. A sterilized soil sample (≈ 300 g) was also prepared as controls to check the chemical degradation of azoxystrobin as indicated by Marín-Benito et al. (2012b). Soil samples for microbiological control were prepared by adding only sterile UHQ water. Soil samples were taken at day 0 for fungicide analysis and thereafter repeatedly at different time intervals (up to 378 days).

2.4. Fungicide extraction and analysis

Samples of moist soil (5 g) were taken by duplicate from each

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